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(54) Ionic compounds and catalyst for olefin polymerisation using the compounds

Ionische Verbindungen und Katalysator für die Olefinpolymerisation, der die Verbindungen verwendet

Composés ioniques et catalyseurs destinés à polymériser les oléfines au moyen de ces composés

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US-A- 5 198 401

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(73) Proprietor: **SHOWA DENKO KABUSHIKI KAISHA**
Minato-ku, Tokyo 105-0011 (JP)

(72) Inventors:
• **HINOKUMA, Shinji**, Oita Research Cntr
Oita-shi, Oita 870-01 (JP)
• **MIYAKE, Shigenobu**, Oita Research Cntr
Oita-shi, Oita 870-01 (JP)
• **ONO, Michio**, Oita Research Cntr
Oita-shi, Oita 870-01 (JP)
• **INAZAWA, Shintaro**, Oita Research Cntr
Oita-shi, Oita 870-01 (JP)

(74) Representative: **Kirkham, Nicholas Andrew**
Graham Watt & Co.,
Riverhead
Sevenoaks, Kent TN13 2BN (GB)

- **JOURNAL OF ORGANOMETALLIC CHEMISTRY**, 27 January 1987, Vol. 319, No. 3, **PEKKA SORMUNEN et al.**, "Modification of Olefin Polymerization Catalysts II", pages 327-332.
- **CHEMICAL ABSTRACTS**, Vol. 89, No. 13, 25 September 1978, (Columbus, Ohio, USA), page 962, Abstract No. 109944z; & **DD-A, 128 638 (AKADEMIE DER WISSENSCHAFTEN DER DDR)** 30.11.77.
- **CHEMICAL ABSTRACTS**, Vol. 92, No. 21, 26 May 1980, (Columbus, Ohio, USA), page 664, Abstract No. 181249f, **BEACHLEY O.T. Jr. et al.**, "Preparation and Properties of [(Trimethylsilyl)methyl]Gallium(III) Compounds"; & **INORG. CHEM.**, 1980, 19(4), 1021-5 (Eng).
- **Journal of Organometallic Chemistry**, 319 (1987), 327-332, **Sormunen et al.**, 'Modification of Olefin Polymerization Catalysts. II. A 29Si NMR Study on the Complexation of Silyl Ethers with Triethylaluminum'
- **Inorganic Chemistry** 1980, 19, 1021-1025, **Beachley et al.**, 'Preparation and properties of ((Trimethylsilyl)methyl)gallium (III) compounds'

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Description

[0001] This invention relates to ionic compounds, method for preparing the same, olefin polymerization catalyst components containing the same, and olefin polymerization catalyst compositions containing the components.

[0002] Heretofore, there has been known a method of producing olefins using a metallocene compound and methylaluminoxane as a catalyst in a homogeneous system. For example, Japanese Patent Application Laid-open No. 19309/1983 discloses a method of producing ethylene homopolymers and ethylene/C₃-C₁₂ α -olefin copolymers using biscyclopentadienyl zirconium dichloride and a linear or cyclic methylaluminoxane as a catalyst. Japanese Patent Application Laid-open No. 130314/1986 discloses a method of producing stereoregular polypropylenes using a catalyst comprising a zirconium compound containing as a ligand a compound having two indenyl groups intervened with an ethylene group and aluminoxane. Also, Japanese Patent Application Laid-open No. 41303/1990 discloses a method of producing poly- α -olefins having a good syndiotacticity.

[0003] There have also been proposed catalyst composition systems that use no aluminoxane cocatalyst. Taube et al. performed polymerization of ethylene using a metallocene represented by [Cp₂TiMe(THF)]⁺[BPh₄]⁻ (Cp: a cyclopentadienyl group, Me: a methyl group, THF: a tetrahydrofuran group, Ph: a phenyl group) (J. Organometall. Chem., 347, C9 (1988)). In J. Am. Chem. Soc., 109, 4111 (1987), Jordan et al. reported that a zirconium complex represented by [Cp₂ZrR(L)]⁺ (Cp: a cyclopentadienyl group, R: a methyl group or a benzyl group, L: a Lewis base) functions as a catalyst for polymerization of ethylene. Japanese Patent Application Laid-open Nos. 501950/1989 and 502036/1989 disclose methods of polymerizing olefins using catalysts comprising a cyclopentadienyl metal compound and an ionic compound which can stabilize the cyclopentadienyl metal cations. Zambelli et al. reported that use of a catalyst which comprises a zirconium compound having a cyclopentadiene derivative as a ligand, trimethylaluminum and fluorodimethylaluminum enables production of isotactic polypropylenes (Macromolecules, 22, 2186 (1989)).

[0004] However, use of the above-described catalysts, when used in slurry system processes or gaseous system processes, caused problems since the polymer is produced in the form of fine powder having a low bulk density and, hence, it is difficult to handle and the polymer produced attaches to an inner wall of the reactor.

[0005] In order to solve these problems, various proposals have been made to have the catalysts carried on solid carriers.

[0006] For example, Japanese Patent Application Laid-open Nos. 108610/1986, 296008/1986, 280703/1988, 22804/1988, 51405/1988, 51407/1988, 55403/1988, 61010/1988, 248803/1988, 100808/1992, 74412/1991, 709/1991 and 7306/1992 disclose methods of producing olefins using solid catalysts comprising inorganic metal oxides such as silica, alumina, silica-alumina and the like having carried thereon metallocene compounds and methylaluminoxane, respectively.

[0007] Japanese Patent Application Laid-open Nos. 6003/1989, 6004/1989, 6005/1989, 11104/1989 and 11105/1989 disclose methods in which catalysts are used that comprise a metallocene compound and aluminoxane carried on an organometallic magnesium compound.

[0008] Also, Japanese Patent Application Laid-open Nos. 260903/1988, 31403/1992 and 74411/1991 disclose polymerization methods in which catalysts are used that comprise a metallocene compound and aluminoxane carried on polymer such as polyethylene, polystyrene, respectively.

[0009] Japanese Patent Application Laid-open Nos. 276805/1986 and 74415/1991 disclose polymerization methods using a metal oxide and a metallocene compound having carried thereon only methylaluminoxane.

[0010] Further, Japanese Patent Application Laid-open Nos. 259004/1989, 259005/1989, 56928/1994 and 56929/1994 disclose methods in which catalysts are used that comprise a metallocene compound having a special ligand, carried on a porous metal oxide carrier, such as silica. On the other hand, Japanese Patent Application Laid-open No. 234405/1992 discloses a method of solidifying a catalyst using a cyclopentadienyl group bonded to a poly (halogenated methylstyrene) to form a complex with the catalyst.

[0011] Studies have been made in order to obtain solidified catalysts for catalyst systems using no aluminoxane cocatalyst. For example, Japanese Patent Application Laid-open Nos. 234709/1991, 247128/1993, 239138/1993, 148316/1993, 155926/1993 and 502906/1993 disclose methods in which catalysts are used that comprise a cation type metallocene compound reacted with a non-coordinating boron compound carried on an inorganic metal compound such as silica. However, in these carrying methods, the boron compound is not bonded to the carrier so that upon polymerization, activated species come off from the surface of the carrier, causing the resulting resin to attach to the reactor.

[0012] DD-A-128 638 discloses the preparation of trialkylsilylmethyltitanium chloride compounds using titanium tetrachloride. US-A-5 198 401 discloses the preparation of ionic catalyst compositions by combining two components.

[0013] The first component is a bis(cyclopentadienyl) Group 4 metal complex. The second component comprises a cation which will irreversibly react with the Group 4 metal complex and a non-coordinating anion. These ionic catalyst compositions can be used to polymerize α -olefins, diolefins and/or acetylenically unsaturated monomers, either alone or in combination, to polymers and copolymers.

[0014] Further, Japanese Patent Application Laid-open No. 501573/1995 (WO 93/11172) discloses a method involving use of an ion-activated transition metal catalyst composition useful for the polymerization of olefins, the composition comprising a core portion and a polyanionic transient metal catalyst component, i.e., a polyanion portion comprising a plurality of metal- or metalloid atom-containing non-coordinating anionic group chemically bonded to the core portion as a side chain. Here, the core portion is comprised by (1) an oligomer of a salt of a polymerizable anionic portion containing a metalloid atom and an organic cation prepared in the presence of a metallocene, (2) crosslinked particles of a polymer such as a styrene based polymer, or (3) inorganic particles such as those of glass, silica, metal, etc. In the above-described publication, there are described examples which used the former two ((1), (2)) as the core and confirmed their effect as a polymerization catalyst. However, when the present inventors tested the examples, the method was insufficient in either one of the activity of olefin polymerization, powder characteristics of the resulting polyolefins, and attachment of the polymers to the reactor.

[0015] Therefore, an object of this invention is to provide a carrier-supported olefin polymerization catalyst which is excellent in the activity of catalyst and has solved the problem involved in the conventional methods that the resulting polymer attaches to the wall of the reactor and to provide an ionic compound for use therein.

SUMMARY OF THE INVENTION

[0016] The present inventors have made intensive research in order to solve the above-described problems and as a result have discovered a Group 13 element (after the 1990 Rule for Nomenclature of Inorganic Compounds)-containing ionic compound having a functional group which is bondable to a carrier and found that highly active polymer which does not attach to the inner wall of the reactor can be obtained by performing polymerization of olefins using an olefin polymerization catalyst comprising a carrier to which the ionic compound is chemically bonded, an organic metal and a transition metal compound belonging to the Group 4, 5, or 6 of the periodic table.

[0017] That is, this invention provides the ionic compounds, methods of producing the same, catalyst components for olefin polymerization catalysts, and olefin polymerization catalysts containing the components described below.

1) Ionic compounds represented by general formula (I) below:



(wherein, M is an element belonging to the Group 13;

R¹, R² and R³, which may be the same or different, represent each a C₁-C₂₀ hydrocarbyl group, a substituted hydrocarbyl group, an alkoxide group, or a halogen atom;

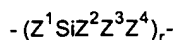
R⁴ represents a C₁-C₂₀ alkylene group, a substituted alkylene group, a substituted phenylene group, a silanylene group, a substituted silanylene group, a silalkylene group, a substituted silalkylene group, an oxasilanylene group, a substituted oxasilanylene group, or an oxasilalkylene group, with k, l and m being each 0 or an integer of 1 to 3, and n being an integer of 1 to 4 such that k + l + m + n = 4;

L is a group represented by general formula (II) or (III) below and is chemically bonded to R⁴:



(wherein R⁵ to R¹², which may be the same or different, represent each a C₁-C₂₀ hydrocarbyl group, a substituted hydrocarbyl group, an alkoxide group, or a halogen atom, with at least one of R⁵, R⁶ and R⁷ and at least one of R⁸, R⁹ and R¹⁰ being a halogen atom;

Y is -O-, a C₁-C₂₀ alkylene group, a substituted alkylene group, a phenylene group, a substituted phenylene group, or a group represented by formula:



(wherein Z¹ and Z⁴, which may be the same or different, represent each an alkylene group, a substituted

alkylene group, a phenylene group, a substituted phenylene group, -O-, an oxyalkylene group, a substituted oxyalkylene group, an oxyphenylene group, or a substituted oxyphenylene group; Z² and Z³, which may be the same or different, represent each a hydrogen atom, an alkyl group, a substituted alkyl group, a phenyl group, or a substituted phenyl group; r is an integer of at least 1);
 5 when n is 2 or more, each R⁴-L may be the same or different;
 D is a monovalent cation selected from the group consisting of carbonium, anilinium, ammonium, ferrocenium, phosphonium, sodium, potassium, or lithium).

2) The ionic compound described in 1) above, wherein n is 1.

3) The ionic compound described in 1) above, wherein M is boron.

4) The ionic compound described in 1) above, wherein R¹, R² and R³ are each a pentafluorophenyl group.

5) The ionic compound described in 1) above, wherein L is a halogenated silyl group, a halogenated substituted silyl group, a halogenated silalkyl group, a halogenated substituted silalkyl group, a halogenated oxasilyl group, a halogenated substituted oxasilyl group, or a halogenated oxasilalkyl group.

6) The ionic compound described in 1) above, wherein R⁴ is a substituted phenylene group.

7) The ionic compound described in 6) above, wherein R⁴ is a 2,3,5,6-tetrafluorophenylene group.

8) The ionic compound described in 1) above, wherein L is a trichlorosilyl group, a methylchlorosilyl group, or a dimethylchlorosilyl group.

9) The ionic compound described in 1) above, wherein D is an anilinium ion.

10) A method of producing ionic compounds represented by general formula (I)



(wherein the symbols in the formula have the same meanings as in 1) above) comprising using components represented by (1) to (4) below:

(1) A compound represented by general formula (IV):



(wherein X¹ and X² independently represent a hydrogen atom or a bromine atom, and R⁴ has the same meaning as in 1) above);

(2) A compound represented by general formula (V):



(wherein M is an element belonging to the Group 13; and R¹, R² and R³, which may be the same or different, represent each a C₁-C₂₀ hydrocarbyl group, substituted hydrocarbyl group, or alkoxide group or a halogen atom);

(3) A compound represented by general formula (VI) or (VII):



(wherein R⁵ to R¹⁴ have the same meanings as in 1) above); and

(4) A halide of a monovalent cation.

11) A method of producing ionic compounds represented by general formula (I)



(wherein the symbols in the formula have the same meanings as in 1) above) comprising the steps of:

(a) substituting bromine or hydrogen a compound represented by general formula (IV)



(wherein the symbols in the formula have the same meanings as in 10) above) by lithium with an organic lithium to obtain a lithium substituted compound;

(b) reacting the lithium substituted compound with a Group 13 element containing compound represented by general formula (V)

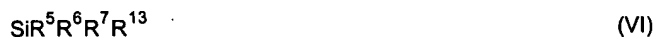


(wherein the symbols in the formula have the same meanings as in 10) above) to obtain a lithium compound represented by general formula (VIII)



(wherein A is hydrogen or bromine chemically bonded to R^4);

(c) lithionating the compound of general formula (VIII) with an organic lithium and then reacting with a silicon compound represented by general formula (VI) or (VII) below:



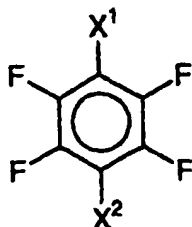
(wherein the symbols in the formulae have the same meanings as in 10) above) to obtain a compound represented by general formula (IX)



(wherein L has the same meaning as in 1) above and other symbols in the formula have the same meanings as above); and

(d) reacting the compound of general formula (IX) with a halide of a monovalent cation.

12) The method of producing the ionic compounds as described in 10) or 11) above, wherein the compound of general formula (IV) is a compound represented by general formula (IVa)



(IVa)

(wherein the symbols in the formula have the same meanings as in 10) above).

13) A Catalyst component for olefin polymerization comprising the ionic compound described in 1) above chemically

bonded to a carrier.

14) The catalyst component as described in 13) above,
wherein the carrier is a solid having a functional group represented by general formula (X)

-OR

(X)

(wherein R is a hydrogen atom, a C₁-C₂₀ alkyl group, alkyl metal or amine).

15) The catalyst component as described in 14) above,
wherein the carrier is a solid having a hydroxyl group.

16) The catalyst component as described in 13) above,
wherein the carrier is silica, alumina or mixtures thereof.

17) A catalyst for olefin polymerization comprising the following as essential components:

- (a) the catalyst component for olefin polymerization as described in 13) above,
- (b) an organometal compound, and
- (c) a Group 4, 5 or 6 transition metal compound.

18) The catalyst as described in 17) above, wherein the Group 4, 5 or 6 transition metal compound is metallocene.

DETAILED DESCRIPTION OF THE INVENTION

[0018] Hereafter, this invention will be described in detail.

[IONIC COMPOUND]

[0019] The ionic compound which can be used in this invention includes those compounds represented by the following general formula (I);



wherein M is a Group 13 metal. Preferred examples of M include boron and aluminum, with boron being particularly preferred.

[0020] R¹, R² and R³ (each bonded to M), which may be the same or different, are selected from a C₁-C₂₀ hydrocarbonyl group, substituted hydrocarbonyl group, or alkoxide group or a halogen atom. These are preferably a C₁-C₂₀ aromatic hydrocarbonyl group, halogen-substituted aromatic hydrocarbonyl group, or halogenated hydrocarbon-substituted aromatic hydrocarbonyl group, and more preferably a phenyl group, a 2-fluorophenyl group, a 3-fluorophenyl group, a 4-fluorophenyl group, a 2,4-difluorophenyl group, a 2,5-difluorophenyl group, a 3,5-difluorophenyl group, a 3,4,5-trifluorophenyl group, a 2,3,4,5-tetrafluorophenyl group, a pentafluorophenyl group, or a 3,5-bis(trifluoromethyl)phenyl group, with a pentafluorophenyl group being most preferred.

[0021] k, l and m are each 0 or an integer of 1 to 3, provided that they have a relation with n described below such that k + l + m + n = 4.

[0022] R⁴ (bonded to M) represents a C₁-C₂₀ alkylene group, substituted alkylene group, substituted phenylene group, silanylene group, substituted silanylene group, silalkylene group, substituted silalkylene group, oxasilanylene group, substituted oxasilanylene group, or oxasilalkylene group.

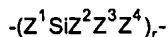
[0023] The present inventors confirmed that the substituent groups of particularly the substituted phenylene group represented by R⁴ give important influence to the activity of the catalyst and the properties of the resulting polymer and that preferred substituents for phenylene are halogens and halogenated hydrocarbons. Preferred substituted phenylene groups include 2-fluorophenyl group, 3-fluorophenyl group, 2,5-difluorophenyl group, 3,5-difluorophenyl group, 2,3,5-trifluorophenyl group, 3,5-bis (trifluoromethyl)phenylene group, 2,3,5,6-tetrafluorophenyl group, among which 2,3,5,6-tetrafluorophenyl group is particularly preferred.

[0024] L (bonded to R⁴) is a carrier bondable functional group. The carrier bondable functional group is a functional group which can form a chemical bond with a surface of a carrier. For example, when there is a hydroxyl group on the surface of a carrier, a compound having a chlorosilyl group can form a silicon-oxygen bond by a reaction with the hydroxyl group. In this case, the chlorosilyl group is a carrier bondable functional group. More specifically, the carrier bondable functional group includes those functional groups represented by general formulae (II) or (III) below:



(wherein R^5 to R^{12} (each bonded to Si), which may be the same or different, represent each a C_1 - C_{20} hydrocarbyl group, substituted hydrocarbyl group, or alkoxide group, or a halogen atom, with at least one of R^5 , R^6 and R^7 and at least one of R^8 , R^9 and R^{10} being a halogen atom;

[0025] Y is -O-, a C_1 - C_{20} alkylene group, substituted alkylene group, phenylene group, substituted phenylene group, or a group represented by formula:



(wherein Z^1 and Z^4 , which may be the same or different, represent each an alkylene group, a substituted alkylene group, a phenylene group, a substituted phenylene group, -O-, an oxyalkylene group, a substituted oxyalkylene group, an oxyphenylene group, or a substituted oxyphenylene group; Z^2 and Z^3 , which may be the same or different, represent each a hydrogen atom, an alkyl group, a substituted alkyl group, a phenyl group, or a substituted phenyl group; r is an integer of at least 1).

[0026] Specific examples of L include a trichlorosilyl group, a methyldichlorosilyl group, a dimethylchlorosilyl group, an ethyldichlorosilyl group, a diethylchlorosilyl group, a phenyldichlorosilyl group, a diphenylchlorosilyl group, a trimethoxysilyl group, a methyldimethoxysilyl group, a dimethylchlorosilyl group, an ethyldimethoxysilyl group, a diethylmethoxysilyl group, a triethoxysilyl group, a methyldiethoxysilyl group, a dimethylethoxysilyl group, an ethyldiethoxysilyl group, a diethylethoxysilyl group, a phenyldiethoxysilyl group, a diphenylethoxysilyl group, a trihydroxysilyl group, a dihydroxyphenylsilyl group, a 2-(dimethylchlorosilyl)ethyldimethylsilyl group, a 6-(dimethylchlorosilyl)hexyldimethylchlorosilyl group, a 8-(dimethylchlorosilyl)octyldimethylchlorosilyl group, a 2(trichlorosilyl)ethyldichlorosilyl group, a 6-(trichlorosilyl)hexyldichlorosilyl group, a 8-(trichlorosilyl)octyldichlorosilyl group, etc. Of these, a trichlorosilyl group, a trimethoxysilyl group, triethoxysilyl group, a trihydroxysilyl group, a dimethylchlorosilyl group, a dimethylmethoxysilyl group, a dimethylethoxysilyl group, a dimethylhydroxysilyl group, a methyldichlorosilyl group, a methyldimethoxysilyl group, a methyldiethoxysilyl group, and a methyldihydroxysilyl group are preferred, with a trichlorosilyl group, a methyldichlorosilyl group, and a dimethylchlorosilyl group being particularly preferred.

[0027] n is an integer of 1 to 4, and when n is 2 or more, R^4 -L's may be a combination of different groups.

[0028] D represents a monovalent cation, which means carbonium, anilinium, ammonium, ferrocenium, phosphonium, sodium, potassium, lithium, etc.

[0029] Specific examples of D include trimethylammonium, triethylammonium, tripropylammonium, tributylammonium, N,N-dimethylammonium, N,N-diethylammonium, N,N-2,4,5-pentamethylanilinium, triphenylphosphonium, tri(p-tolyl)phosphonium, triphenylcarbonium, etc. Of these, N,N-dimethylanilinium and triphenylcarbonium are preferred.

[0030] Specific examples of such an ionic compound include N,N-Dimethylanilinium salts, for example, the following compounds.

N,N-Dimethylanilinium tris(pentafluorophenyl) p-trichlorosilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-methyldichlorosilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-dimethylchlorosilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris (pentafluorophenyl) p-trimethoxysilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-dimethoxymethylsilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-methoxydimethylsilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-triethoxysilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-diethoxymethylsilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-ethoxydimethylsilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-trihydroxysilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-dihydroxymethylsilyltetrafluorophenylborate,
 N,N-Dimethylanilinium tris(pentafluorophenyl) p-hydroxydimethylsilyltetrafluorophenylborate,
 Triphenylcarbonium tris(pentafluorophenyl) p-trichlorosilyltetrafluorophenylborate,
 Triphenylcarbonium tris (pentafluorophenyl) p-methyldichlorosilyltetrafluorophenylborate,
 Triphenylcarbonium tris (pentafluorophenyl) p-dimethylchlorosilyltetrafluorophenylborate,
 Triphenylcarbonium tris (pentafluorophenyl) p-trimethoxysilyltetrafluorophenylborate,

Triphenylcarbenium tris (pentafluorophenyl) p-dimethoxymethylsilyltetrafluorophenylborate,
 Triphenylcarbenium tris (pentafluorophenyl) p-methoxydimethylsilyltetrafluorophenylborate,
 Triphenylcarbenium tris(pentafluorophenyl) p-triethoxysilyltetrafluorophenylborate,
 Triphenylcarbenium tris (pentafluorophenyl) p-diethoxymethylsilyltetrafluorophenylborate,
 5 Triphenylcarbenium tris(pentafluorophenyl) p-ethoxydimethylsilyltetrafluorophenylborate,
 Triphenylcarbenium tris(pentafluorophenyl) p-trihydroxysilyltetrafluorophenylborate,
 Triphenylcarbenium tris(pentafluorophenyl) p-dihydroxymethylsilyltetrafluorophenylborate,
 Triphenylcarbenium tris(pentafluorophenyl) p-hydroxydimethylsilyltetrafluorophenylborate,

10 [0031] Of these ionic compounds, preferred are N,N-dimethylanilinium tris (pentafluorophenyl) 4-(trichlorosilyl)-
 2,3,5,6-tetrafluorophenylborate, N,N-dimethylanilinium tris(pentafluorophenyl) 4-(methyldichlorosilyl)-
 2,3,5,6-tetrafluorophenylborate, and N,N-dimethylanilinium tris (pentafluorophenyl) 4-(dimethylchlorosilyl)-
 2,3,5,6-tetrafluorophenylborate.

15 [Production Method for Ionic Compounds]

[0032] The ionic compounds of this invention can be produced using the following compounds.

(1) A compound represented by general formula (IV):



(wherein X¹ and X² independently represent a hydrogen atom or a bromine atom, and R⁴ has the same meaning
 25 as above);

(2) A compound represented by general formula (V):



(wherein M, R¹, R² and R³ have the same meanings as above);

(3) A compound represented by general formula (VI) or (VII):



(wherein R⁵ to R¹² have the same meanings as above, and R¹³ and R¹⁴ independently represent a C₁-C₂₀ hydro-
 carbonyl group, substituted hydrocarbonyl group, alkoxide group, or a halogen atom); and

(4) A halide of a monovalent cation.

45 [0033] As the reaction process for producing these compounds, there may be considered various reaction proce-
 dures. However, any process may be used as far as there can finally be obtained compounds represented by general
 formula (I)



(wherein the symbols in the formula have the same meanings as above).

55 [0034] As the most typical process, there can be cited a reaction process including the following steps (a) to (d). This
 reaction process, described briefly, comprises the steps of activating a compound containing a R⁴ group (first step),
 bonding the compound with a compound containing a Group 13 element (M) (second step), introducing a carrier bond-
 able functional group to the resulting compound (L) (third step), and then forming a salt with a cation [D]⁺ (fourth step).

[0035] Hereafter, each of the steps is described in detail.

(a) First step:

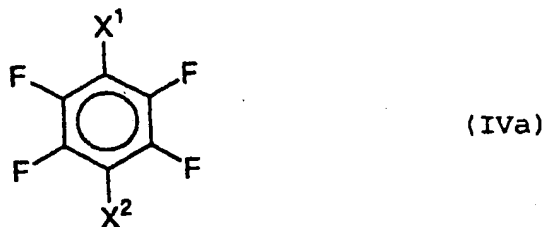
[0036] Bromines or hydrogens in a compound represented by general formula (IV)



(wherein X^1 and X^2 are bromine or hydrogen provided that for compounds having low acidities, at least one of the symbols must be bromine) are substituted by lithium (lithiated) with an organic lithium.

[0037] The lithiation can be performed by a conventional method. More specifically, the compound of formula (IV) above and an organic lithium compound are mixed in a nonreactive solvent to allow reaction therebetween.

[0038] The compound of formula (IV) is selected depending on final target compound and is preferably a halogenated aryl or a halogenated hydrocarbon-substituted aryl as described above. Particularly preferred are tetrafluoro-mono- (or - di)-bromobenzene represented by the following formula (IVa)



[0039] Any organic lithium compounds may be used without limitation as far as they are commonly used for lithiation. For example, there can be cited n-butyllithium, t-butyllithium, and phenyllithium. It is preferred that these are dissolved in an inert solvent such as hexane before they can be subjected to lithiation. Preferred reaction solvents include ether based solvents such as diethyl ether and tetrahydrofuran.

[0040] The reaction between the compound of formula (IV) and the organic lithium compound can be performed in proportions of about 1:1 to 1:10 (by mole), and preferably 1:1 (by mole). The reaction temperature is -100 to 0°C, and preferably -80 to -20°C. It is preferred to allow the reaction to proceed gently by adding the organic lithium compound in portions. The reaction time lasts preferably for 30 minutes or longer.

(b) Second step: Formation of a salt containing a Group 13 element-containing compound

[0041] The lithium substituted compound obtained in the first step is reacted with a Group 13 element-containing compound represented by general formula (V)



(wherein the symbols in the formula have the same meanings as above) to obtain a lithium compound represented by general formula (VIII)



(wherein A is hydrogen or bromine).

[0042] In the formula (V) above, M is a Group 13 element, among which preferred are boron and aluminum, with particularly preferred being boron. In the same formula, R^1 , R^2 and R^3 , which may be the same or different, may be a C1-C20 hydrocarbonyl group, substituted hydrocarbonyl group, or alkoxide group, or a halogen atom. Preferably, they represent a phenyl group, a 2-fluorophenyl group, a 3-fluorophenyl group, a 4-fluorophenyl group, a 2,4-difluorophenyl group, a 2,5-difluorophenyl group, a 3,5-difluorophenyl group, a 3,4,5-trifluorophenyl group, a 2,3,4,5-tetrafluorophenyl group, a pentafluorophenyl group, a 3,5-bis(trifluoromethyl)phenyl group, with a pentafluorophenyl group being particularly preferred.

[0043] This reaction can be performed by mixing the lithionated compound obtained in the first step with a solution of the compound of general formula (V) in a solvent in proportions such that the molar ratio of the lithionated compound to the compound of general formula (V) is 1.0 or more. Preferred mixing ratio is 1.0 to 2.0. When the lithionated compound is added, it is preferred that the temperature of the solution is kept low. More specifically, the temperature is -100 to 50°C, and preferably -20 to 30°C. As for the solvent for dissolving the compound of general formula (V), there can be used liquids which are nonreactive with the lithionated compound and the compound of general formula (V). Preferred solvent includes C₁₀-C₂₀ hydrocarbons and ethers. Particularly preferred ones are toluene, heptane, decane, isoparaffin, diethyl ether, and tetrahydrofuran. The reaction time is preferably 30 minutes or longer. It is preferred that after the reaction, unreacted materials are removed by washing or the like. The compound of general formula (V) may be used in the state where a solvent such as ether is added.

(c) Third step: Introduction of Carrier Bondable Functional Group

[0044] The compound of general formula (VIII) is lithionated with an organic lithium and then reacted with a silicon compound represented by general formula (VI) or (VII) below



(wherein R⁵ to R¹² have the same meanings as above, and R¹³ and R¹⁴ independently represent a C₁-C₂₀ hydrocarbyl group, substituted hydrocarbyl group, alkoxide group, or a halogen atom) to obtain a compound represented by general formula (IX)



(wherein the symbols in the formula have the same meanings as above).

[0045] The mixing ratio of the compound of general formula (VIII) and the silicon compound, as molar ratio of the latter to the former, is 1 fold to 100 folds, preferably 2 folds to 20 folds. Stirring is performed for a certain time at room temperature to complete the reaction. Thereafter, a hydrocarbon having a boiling point higher than the solvent is added to the reaction mixture, the reaction solvent is distilled off, and the hydrocarbon solvent is removed. After washing with the hydrocarbon solvent, the washing solvent is removed. The compound thus obtained is dissolved in a solvent such as dichloromethane.

(d) Fourth step: Cation Exchange Reaction

[0046] Finally, the compound of general formula (IX) is reacted with a halide of a cation D to yield a target ionic compound represented by general formula (I)



(wherein the symbols in the formula have the same meanings as above).

[0047] The halide of D is a compound which generates a monovalent cation D when reacted with the compound of general formula (IX) above. More specifically, the halide of D includes trimethylamine hydrochloride, triethylamine hydrochloride, tripropylamine hydrochloride, tributylamine hydrochloride, N,N-dimethylaniline hydrochloride, N,N-diethylamine hydrochloride, N,N-2,4,5-pentamethylaniline hydrochloride, triphenylphosphine hydrochloride, triphenylchloromethane and the like. Of these, preferred are N,N-dimethylaniline hydrochloride and triphenylchloromethane.

[0048] The reaction can be performed at -100 to 200°C, preferably at 20 to 150°C. The reaction time is preferably 1 hour or longer. Lithium halide precipitates and removal of which yields the target compound. The compound can be used in a state of solution or in a state of solid after drying.

[Catalyst Component for Olefin Polymerization]

[0049] Next, description will be made of the catalyst component for olefin polymerization, comprising the ionic compound of this invention chemically bonded to a carrier.

[0050] In this invention, by the term "chemical bonds" is meant covalent bonds, ionic bonds, metal bonds, and coordination bonds.

(1) Carrier

[0051] In this invention, by the term "carrier" is meant a solid which has a surface with which a plurality of the ionic compounds of this invention can form chemical bonds, the surface having an area and properties such that upon polymerization reaction, a plurality of polymerization active sites can be formed.

[0052] As the carrier, there can be used inorganic oxides, inorganic chlorides, inorganic hydroxides, organic high molecular weight compounds. More specifically, there can be used one or more members selected from the group consisting of inorganic compounds such as silica, alumina, silica-alumina, magnesia, titania, zirconia, and calcia, polymers of vinyl chloride, vinyl alcohol, methacrylic acid, acrylic acid and the like, or copolymers thereof with styrene, divinylbenzene or the like, and homopolymer or copolymer of α -olefins chemically modified to have a functional group which can react with a carrier bondable functional group of the ionic compound to form a chemical bond to the ionic compound. It is preferred to use one or more of silica, alumina or silica-alumina.

[0053] Among the carriers, preferred are those having a functional group represented by general formula (X)



(X)

on the surface of the carrier.

[0054] In the above formula, G represents hydrogen, a C_1 - C_{20} alkyl group, alkylmetal or amine. Of these, preferred are hydrogen, a methyl group, an ethyl group, sodium and lithium, with hydrogen being most preferred.

[0055] The carrier in this invention requires large surface area and pore diameter and functional groups on the surface for chemically modifying the surface with an ionic compound. Further, the ionic compound borne from polymerization active species by forming ion pairs with the transient metal compound so that the carrier must have space for forming such ion pairs. In addition, polymerization activity per catalyst is higher, the larger the amount of the ion pair formed on the carrier and, hence, it is desired that the carrier have a large surface area and a large average pore diameter. As the carrier in this invention, it is preferred to use fine particles having an average particle diameter of 5 to 200 μm , a specific surface area of 100 to 1,000 m^2/g , and an average pore diameter of $2 \times 10^{-9}\text{m}$ (20 Å) or more.

(2) Reaction of the Ionic Compound with the Carrier

[0056] The reaction between the carrier in this invention and the ionic compound having a carrier bondable functional group can be performed by various methods. Generally, the reaction is performed in organic solvents. More specifically, there can be used aliphatic hydrocarbons such as pentane, hexane, heptane, octane, nonane, and decane, alicyclic hydrocarbons such as methylcyclopentane, cyclopentane, and cyclooctane, aromatic hydrocarbons such as benzene, toluene, xylene, cumene, and cymene, aliphatic halogenated hydrocarbons such as chloroform and dichloromethane, aromatic halogenated hydrocarbons such as chlorobenzene, and dichlorobenzene, alcohols such as methanol and ethanol, ethers such as diethyl ether and tetrahydrofuran, and the like.

[0057] Although the reaction between the carrier and the ionic compound having a carrier bondable functional group can be under any conditions as far as the target bond is formed, generally the following conditions are desirable.

[0058] The reaction temperature is usually -70°C to 200°C , and preferably 0°C to 150°C . It is preferred that the reaction between the ionic compound having a carrier bondable functionality with the functional group on the surface of the carrier proceed sufficiently.

[0059] The reaction time may vary depending on the concentration, temperature and other conditions and is not limited generally. Usually, the reaction can proceed for 1 to 50 hours.

[0060] The reaction for bonding can be carried out at a concentration of the ionic compound in the reaction solvent being 1 to 10,000 ppm and at a concentration of the carrier being 1 to 50 wt. %.

[0061] There is no particular limitation in the proportion of amounts of the ionic compound having a carrier bondable functional group and of the carrier. When the number of the functional groups on the surface of the carrier is equal to or greater than the equivalent the carrier bondable functional group, unreacted functional groups on the surface of the carrier will react with the transition metal compound so that there is the possibility that no polymerization active species can be formed. In this case, the reaction with the transition metal compound can be prevented from occurring by

reacting the functional group on the surface of the carrier with a different compound. For example, where the unreacted functional group is a hydroxyl group, treatment of the hydroxyl group with trimethylchlorosilane or the like results in the prevention of the reaction between the transient metal compound and the unreacted functional group on the surface of the carrier.

[0062] The catalyst component for olefin polymerization, the reaction product, is separated from the reaction mixture and the ionic compounds having unreacted carrier bondable functional groups are removed by washing. As the solvent for washing, there can be used the above-described organic solvents. The temperature for washing is -30°C to 120°C, and preferably 0°C to 100°C. Preferably, the washing is continued until the ionic compound having a carrier bondable functional group cannot be detected substantially in the washings. After completion of the washing, the solid component carrying the ionic compounds is dried or may be used in the presence of an organic solvent.

[0063] Formation of chemical bonds between the ionic compound having a carrier bondable functional group and the carrier can be confirmed by determination of a reduction in the amount of the functional group on the carrier as a result of the reaction or by determination of the amount of the compound which is formed as a result of the reaction between the functional group on the surface of the carrier and the carrier bondable functional group of the ionic compound. Alternatively, such is confirmed by detecting IR absorption and NMR peaks of new bonds which are formed as a result of the reaction.

[Catalyst for Olefin Polymerization]

[0064] Next, description will be made of the catalyst for olefin polymerization of this invention.

[0065] The component for olefin polymerization catalyst is characterized by using

- (a) a component of olefin polymerization catalyst in which the ionic compound is chemically bonded to the carrier,
- (b) an organometal compound, and
- (c) a Group 4, 5 or 6 transition metal compound.

[0066] Of these, (a) has already been described above.

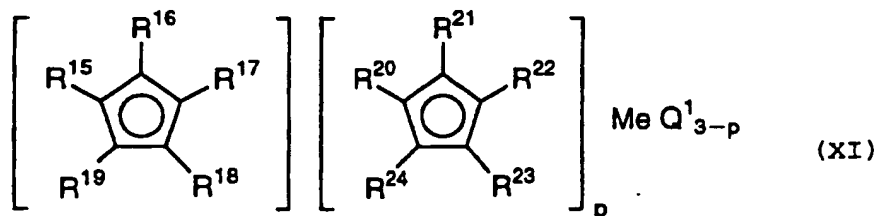
[0067] In the organometal compound (component (b)) which is one of the catalyst component for olefin polymerization in this invention, the metal element is generally lithium, sodium, magnesium, aluminum, tin, zinc or titanium. The organic group which combines with the metal to form an organic metal compound generally includes an alkyl group (C₁-C₁₀), and a phenyl group, a cyclopentadienyl group or derivatives thereof.

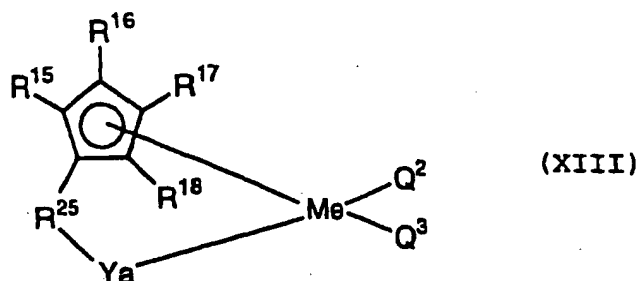
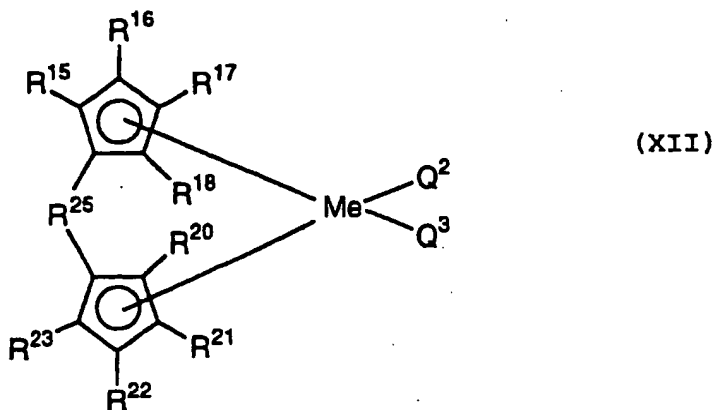
[0068] At least one of the valence of the above-described metal element must be satisfied with the above-described organic group while the rest valences may be satisfied with other atoms or atomic groups. Such atoms or atomic groups may be, for example, a halogen atom, a hydrogen atom, an alkoxy group, etc.

[0069] As such organic compounds, there can be cited, for example, organolithium compounds such as n-butyllithium, t-butyllithium, and phenyllithium, organosodium compounds such as cyclopentadienylsodium and methylsodium, organomagnesium compounds such as butylethylmagnesium, butyloctylmagnesium, ethylmagnesium bromide, and butylmagnesium bromide, organoaluminum compounds such as trimethylaluminum, triethylaluminum, triisobutylaluminum, diethylaluminum halide, diethylaluminum chloride, diethylaluminum bromide, diethylaluminum ethoxide, ethylaluminum sesquichloride, and isobutylaluminum oxane, organotin compounds such as tetraethyltin, tetrabutyltin, tributyltin chloride, and tetraphenyltin, organozinc compounds such as diethylzinc, and dibutylzinc, organotitanium compounds such as dicyclopentadienyltitanium dimethyl. Of these, preferred are organoaluminum compounds and organomagnesium compounds.

[0070] In the catalyst for olefin polymerization of this invention, the organometal compound component may be used as a combination of two or more of them.

[0071] The Group 4, 5 or 6 transition metal compound (component (c)), one of the components for olefin polymerization catalyst, is preferably a transition metal compound represented by general formulae (XI), (XII) or (XIII) below





[wherein R^{15} to R^{24} , which may be the same or different, represent each a hydrogen atom or a hydrocarbonyl group (C_1 - C_{20} alkyl, alkenyl, aryl, alkylaryl, arylalkyl or the like), an alkylsilyl group, an alkylgermyl group, or a 4-6 membered ring having a carbon-carbon bond, which may be the same or different, R^{25} represents a C_1 - C_{20} alkylene group, an alkylgermylene or alkylsilylene,

a plurality of Q^1 s, and Q^2 and Q^3 , which may be the same or different, represent each a C_1 - C_{20} aryl, alkyl, alkenyl, alkylaryl, arylalkyl or the like hydrocarbonyl group, alkoxy, alkoxy, siloxy, hydrogen or halogen, Y_a is an electron donating ligand selected from $-O-$, $-S-$, $-NR^{26}-$, $-PR^{26}-$, or $-OR^{26}$, $-SR^{26}$, $-NR^{26}R^{27}$, and $-PR^{26}R^{27}$ (R^{26} and R^{27} represent each hydrogen or a C_1 - C_{20} alkyl, alkenyl, aryl, alkylaryl, arylalkyl or the like hydrocarbonyl group, or halogenated alkyl or halogenated aryl), Me is a transition metal of Group 3, 4, 5 and 6 of the periodic table, p is 0 or 1].

[0072] In the above formulae, the transition metal elements of Group 4, 5 and 6 of the periodic table (Group being after 1990 Rule of Inorganic Compound Nomenclature) are preferably selected from the transition metal elements of Group 4 of the periodic table, i.e., titanium, zirconium, and hafnium, with particularly preferred being zirconium and hafnium.

[0073] In the above formulae, the hydrocarbonyl group represented by R^{15} to R^{24} include, for example, a methyl group, an ethyl group, a propyl group, a butyl group, an isobutyl group, a t -butyl group, an amyl group, an isoamyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, a cetyl group, a phenyl group, etc., the alkylsilyl group includes a trimethylsilyl group, etc., the alkylgermyl group includes a trimethylgermyl group, etc. As the cyclopentadienyl ligand, there can be cited, for example, alkyl substituted cyclopentadienyl group such as a cyclopentadienyl group, a methylcyclopentadienyl group, an ethylcyclopentadienyl group, an n -butylcyclopentadienyl group, a t -butylcyclopentadienyl group, a trimethylsilylcyclopentadienyl group, a dimethylcyclopentadienyl group, a pentamethylcyclopentadienyl group, and an indenyl group, a fluorenyl group and the like having or not having similar substituents.

[0074] In the above formulae, as the alkylene group represented by R^{25} , there can be cited, for example, a methylene group, an ethylene group, a propylene group, an isopropylidene group, a cyclopentylidene group, a cyclohexylidene group, a tetrahydropyran-4-ylidene group, a diphenylmethylene group, etc.. As the alkylsilylene group, there can be cited, for example, a dimethylsilylene group, a diphenylsilylene group, etc. As the alkylgermylene group, there can be cited, for example, a dimethylgermylene group, a diphenylgermylene group, etc.

[0075] In the above formulae, specific examples of R^{26} and R^{27} in Y_a include a methyl group, an ethyl group, a propyl group, a butyl group, an isobutyl group, a t -butyl group, an amyl group, an isoamyl group, a hexyl group, a heptyl group,

an octyl group, a nonyl group, a decyl group, a cetyl group, a phenyl group, a benzyl group, etc.

[0076] In Ya, ligands of the type -NR²⁶- or -PR²⁶- are preferred.

[0077] Hereafter, specific examples of the transition metal compound represented by general formulae (XI), (XII) or (XIII) when Me is zirconium are exemplified.

[0078] Examples of the transition metal compounds represented by general formula (XI) include:

Biscyclopentadienylzirconium dichloride,
 Bis(methylcyclopentadienyl)zirconium dichloride,
 Bis(n-butylcyclopentadienyl)zirconium dichloride,
 Bis(n-butylcyclopentadienyl)zirconium dimethyl,
 Bis(1,3-dimethylcyclopentadienyl)zirconium dichloride,
 Bis(pentamethylcyclopentadienyl)zirconium dichloride,
 (Cyclopentadienyl)(methylcyclopentadienyl)zirconium dichloride,
 (Cyclopentadienyl)(n-butylcyclopentadienyl) zirconium dichloride,
 (Cyclopentadienyl) (indenyl) zirconium dichloride,
 (Cyclopentadienyl)(fluorenyl)zirconium dichloride,
 Cyclopentadienylzirconium trichloride,
 Cyclopentadienylzirconium trimethyl,
 Pentamethylcyclopentadienylzirconium trichloride,
 Pentamethylcyclopentadienylzirconium trimethyl, etc.

[0079] Examples of the transition metal compound represented by general formula (XII) include:

Dimethylsilylenebis (methylcyclopentadienyl)zirconium dichloride,
 Isopropylidenebis(methylcyclopentadienyl) zirconium dichloride,
 Ethylenebis(indenyl)zirconium dichloride,
 Ethylenebis (4,5,6,7-tetrahydro-1-indenyl)zirconium dichloride,
 Isopropylidene (cyclopentadienyl) (fluorenyl) zirconium dichloride,
 Isopropylidene(cyclopentadienyl)(indenyl)zirconium dichloride,
 Isopropylidene (t-butylcyclopentadienyl)(t-butylindenyl)zirconium dichloride,
 Isopropylidene(t-butylcyclopentadienyl)(t-butylindenyl)zirconiumdimethyl,
 Dimethylsilylene-bis(2-methyl-4-benzoindenyl))-zirconium dichloride,
 Dimethylsilylene-bis(1-(2-methyl-4-phenylindenyl))-zirconium dichloride,
 Dimethylsilylene-bis(1-(2-methyl-4-naphthylindenyl))-zirconium dichloride, etc.

[0080] Examples of the transition metal compound represented by general formula (XIII) include:

Ethylene(t-butylamide)(tetramethylcyclopentadienyl)-zirconium dichloride,
 Ethylene(methylamide)(tetramethylcyclopentadienyl)-zirconium dichloride,
 Dimethylsilylene(t-butylamide)(tetramethyl-cyclopentadienyl) zirconium dichloride,
 Dimethylsilylene(t-butylamide)(tetramethylcyclopentadienyl)zirconium dibenzyl,
 Dimethylsilylene(benzylamide)(tetramethylcyclopentadienyl) zirconium dibenzyl,
 Dimethylsilylene(phenylamide)(tetramethylcyclopentadienyl)zirconium dichloride, etc.

[0081] Herein, the zirconium compounds were exemplified by referring to their specific names. Also, those transition metal compounds in which zirconium is replaced by hafnium or titanium are useful in this invention.

[0082] Regarding the use of the transition metal compounds according to this invention, the above-described transition metal compounds can be used singly or two or more of them may be used in combination.

[0083] Molar ratio of the transition metal compound and the Group 13 element in the components (a), (b) and (c) for olefin polymerization in this invention is such that upon polymerization, the transition metal is 0.01 to 10 times, preferably 0.1 to 1 time, as much as the Group 13 element. The concentration of the transition metal compound upon polymerization can be 0.01 to 100 ppm, preferably 0.1 to 10 ppm.

[0084] The catalyst for olefin polymerization of this invention can be used by contacting (a), (b) and (c) recited in the claims in a solvent or in the presence of a monomer. Methods for contacting the components are not limited particularly. However, a method is preferred in which the component (a) is contacted with the component (b) in an inert solvent and then mixed with the component (c).

[0085] Also, after the components (a), (b) and (c) are contacted, the solvent may be distilled off. Alternatively, the mixture may be washed with a solvent such as a hydrocarbon and can be used as a slurry or the solvent may be

distilled off thereafter.

[0086] The catalyst thus prepared is charged in a polymerization apparatus. In this case, it is preferred that the above-described organometal compound is charged in the polymerization reactor in advance. Particularly when no washing with a hydrocarbon solvent is performed, it is preferred to charge the organometal compound (e.g., organolithium compound) in the polymerization reactor in advance.

[Polymerization of Olefins Using the Catalyst]

[0087] In this invention, methods and conditions of polymerization using the above-described catalyst are not limited particularly and there can be used, for example, solution polymerization methods, melt polymerization methods, slurry polymerization methods, suspension polymerization methods, gas phase polymerization methods and the like. This invention is effective particularly for processes in which polymers are non-homogeneous. More specifically, slurry methods and gas phase methods are exemplified.

[0088] The polymerization temperature is within the ranges of -100 to 300°C, and preferably 0 to 150°C, and the polymerization pressure is within the ranges of atmospheric pressure to 9.8×10^6 Pa (100 kg/cm²), and preferably atmospheric pressure to 4.9×10^6 Pa (50 kg/cm²). The polymerization reaction may be any of batch methods, semi-continuous methods, and continuous methods. Further, multi-step polymerization can also be performed.

[0089] The molecular weight of the resulting polymer can be controlled by use of a chain transfer agent such as hydrogen or polymerization temperature.

[0090] In this invention, as the hydrocarbon solvent which can be used in the polymerization, there can be cited, for example, aliphatic hydrocarbons such as propane, butane, isobutane, pentane, hexane, heptane, octane, nonane, and decane, alicyclic hydrocarbons such as methylcyclopentane, cyclopentane, and cyclooctane, aromatic hydrocarbons such as benzene, toluene, xylene, cumene and cymene. These solvents may be used singly or two or more of them can be used in admixture.

[0091] As the olefin to be used for polymerization, there can be cited, for example, ethylene, propylene, 1-butene, 3-methyl-1-butene, 1-hexene, 4-methyl-1-pentene, 1-octene, 1-decene, 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene, 1-eicosene, cyclopentene, cycloheptene, norbornene, 5-methyl-2-norbornene, tetracyclododecene, etc. Further, there can be cited, for example, styrene, vinylcyclohexane, vinylcyclohexene, divinylbenzene, dienes, etc.

[0092] In this invention, not only homopolymerization of olefins can be performed but also copolymers of, for example, ethylene with propylene, ethylene with 1-butene, and the like can be produced.

[0093] In the polymerization method of this invention, preliminary polymerization can be performed using the catalyst of this invention. There is no particular limitation on the method of the preliminary polymerization but known methods can be used therefor. Olefins to be used for preliminary polymerization are not limited particularly and the above-described olefins can be used. The temperature for preliminary polymerization is usually -20 to 300°C, preferably -10 to 200°C, and more preferably 0 to 100°C. As the solvent, there can be used inert hydrocarbons, aliphatic hydrocarbons, aromatic hydrocarbons, monomers and the like. Preferred are aliphatic hydrocarbons. Also, the preliminary polymerization can be performed without solvents.

[0094] The catalyst containing the ionic compound carrying carrier of this invention has high activity so that polymerization of olefins by a slurry process, a gas phase process or the like using the catalyst gives rise to a polymer having good powder characteristics and attachment of the polymer to the reactor is minimized.

BEST MODE FOR CARRYING OUT THE INVENTION

[0095] Hereafter, this invention will be described in more detail by examples and comparative examples. However, this invention is not limited to the examples. In the following examples, "%" is by weight unless otherwise indicated specifically.

Example 1:

50 Production of Dimethylanilinium 4-trichlorosilyl-2,3,5,6-tetrafluorophenyltris(pentafluorophenyl)borate [Ionic Compound]

[0096] 3.85 Grams (1.86 mmol) of 1-bromo-2,3,5,6-tetrafluorobenzene was dissolved in 50 ml of diethyl ether and cooled in a dry ice/denatured alcohol bath. To the solution was added dropwise 10.5 ml of a solution of 1.6 mol of n-butyllithium/liter of hexane, followed by stirring for 30 minutes to prepare a lithionated product of 1-bromo-2,3,5,6-tetrafluorobenzene (2,3,5,6-tetrafluorophenylithium).

[0097] The reaction mixture containing the above-described lithionated compound was added to 200 ml of a hexane solution containing 5.07 g (9.9 mmol) of tris(pentafluorophenyl)borane, and stirred at 25°C for 20 minutes to precipitate

product (A). The solution layer was removed and vacuum dried after it was washed with hexane.

[0098] 1.66 Grams of the product (A) was dissolved in 10 ml of tetrahydrofuran and cooled in a dry ice/denatured alcohol bath. To this solution was added dropwise 1.5 ml of a solution of 1.6 mol of n-butyllithium/liter of hexane, followed by stirring for 45 minutes. Thereafter, the solution was added to a solution of 2.7 ml of silicon tetrachloride in 10 ml of tetrahydrofuran, and the mixture was stirred at 25°C for 15 minutes. Further, 100 ml of heptane was added and tetrahydrofuran was distilled off. The heptane layer was removed and dried under vacuum after it was washed with hexane. The product was dissolved in 50 ml of dichloromethane and after lithium chloride which precipitated was removed, the product was dried under vacuum to yield 1.65 g of product (B).

[0099] 1.65 Grams of product (B) was dissolved in 30 ml of dichloromethane and 0.31 g of dimethylanilinium chloride was added, and the mixture was stirred at 25°C for 5 minutes. Lithium chloride which precipitated was removed and the product was dried under vacuum to yield 1.7 g of dimethylanilinium 4-trichlorosilyl-2,3,5,6-tetrafluorophenyltris(pentafluorophenyl)borate. Its chemical structure was confirmed by NMR data measured (a part of which is shown below).

¹H-NMR (CD₂Cl₂): δ 3.23(6H), 7.50(2H), 7.57(3H), 11.5(1H);

¹⁹F-NMR (CD₂Cl₂): δ -131.0, -131.6, -132.9, -163.4, -167.3

Example 2:

Production of Dimethylanilinium 4-dimethylchlorosilyl-2,3,5,6-tetrafluorophenyltris(pentafluorophenyl)borate

[Ionic Compound]

[0100] 7.5 Grams (50 mmol) of 1,2,4,5-tetrafluorobenzene was dissolved in 50 ml of diethyl ether and cooled in a dry ice/denatured alcohol bath. To the solution was added dropwise 31.3 ml of a solution of 1.6 mol of n-butyllithium/liter of hexane, followed by stirring for 30 minutes. The resulting solution was added to 883 ml (50 mmol) of 2.9% isoparaffin solution of tris(pentafluorophenyl)borane, and stirred at 25°C for 30 minutes. Upon this, product (C) precipitated. The solution layer was removed and washed with hexane, followed by drying under vacuum. The product (C) was recrystallized from diethyl ether (yield: 34.5 g).

[0101] 8.44 Grams (9.5 mmol) of the product (C) was dissolved in 30 ml of tetrahydrofuran and cooled in a dry ice/denatured alcohol bath. To this solution was added dropwise 6.25 ml of a solution of 1.6 mol of n-butyllithium/liter of hexane, followed by stirring for 45 minutes. The resulting solution was added to 50 ml of tetrahydrofuran having dissolved therein 12 ml (10 mmol) of dimethyldichlorosilane, and stirred at 25°C for 30 minutes. After the stirring, 100 ml of heptane was added and tetrahydrofuran was distilled off. The heptane layer was removed and washed with hexane, followed by drying under vacuum. The product was dissolved in 30 ml of dichloromethane, to which was added 20 ml of a dichloromethane solution of 2.99 g (19.0 mmol) of N,N-dimethylaniline hydrochloride. The solution was stirred for 30 minutes and lithium chloride which precipitated was removed and dried under vacuum to yield 1.6 g of dimethylanilinium 4-dimethylchlorosilyl-2,3,5,6-tetrafluorophenyltris(pentafluorophenyl)borate. Its chemical structure was confirmed by NMR data measured (a part of which is shown below).

¹H-NMR (CD₂Cl₂): δ 0.77(6H), 3.23(6H), 7.50(6H), 7.57(3H), 11.5(1H);

¹⁹F-NMR (CD₂Cl₂): δ -128.6, -129.1, -130.0, -159.8, -163.6.

Example 3:

Production of Dimethylanilinium 4-(8-(dimethylchlorosilyl)-octyldichlorosilyl-2,3,5,6-tetrafluorophenyl)-tris(pentafluorophenyl)borate [Ionic Compound]

[0102] The same procedures as in Example 2 were repeated except that 1,8-bis(dimethylchlorosilyl)octane was used in place of dimethyldichlorosilane to yield 2.3 g of dimethylanilinium 4-(8-(dimethylchlorosilyl)-octyldichlorosilyl)-2,3,5,6-tetrafluorophenyltris(pentafluorophenyl) borate.

Comparative Example 1:

Production of Dimethylanilinium 4-dimethylchlorosilylphenyltris(pentafluorophenyl)borate [Ionic Compound]

[0103] The same procedures as in Example 2 were repeated except that 1,4-dibromobenzene was used in place of 1,2,4,5-tetrafluorobenzene to yield 0.8 g of the target compound, dimethylanilinium 4-dimethylchlorosilylphenyl-tris(pentafluorophenyl)borate.

Example 4:

Preparation of Components for Olefin Polymerization Catalyst from the Compound of Example 1

5 **[0104]** To 30 ml of dichloromethane was added 0.5 g of silica dried at 150°C and at 0.5 mmHg for 4 hours, with adding 6 ml of a dichloromethane solution containing 56 µmol/ml of the ionic compound having a carrier bondable functional group obtained in Example 1 while stirring, and the resulting solution was refluxed for 2 hours. This was washed three times with 20 ml of dichloromethane at 40°C and dried under vacuum.

10 **[0105]** The olefin polymerization catalyst component thus obtained was measured for its boron content by Induction Combined High Frequency Plasma (ICP) spectrophotometry. As a result, it was confirmed that the 0.25 mmol of the ionic compound of Example 1 was carried per g of the catalyst component. After the carrying, the amount of hydroxyl groups on the surface of silica was determined by measuring the amount of ethane which is formed as a result of the reaction between the hydroxyl groups on the surface of silica and triethylaluminum. As a result, the hydroxyl groups of the silica moiety was confirmed to be decreased by about 0.25 mmol/g. From this result, it is understood that sub-

15 substantially all the ionic compounds carried on silica reacted with the hydroxyl groups on the surface of silica. Further, formation of hydrogen chloride upon the reaction between the ionic compounds and silica by contacting the gaseous dichloromethane used in the reaction solvent with water, neutralizing with sodium carbonate and titrating with an aqueous silver nitrate solution using potassium chromate as an indicator.

20 **[0106]** The olefin polymerization catalyst component thus obtained was washed three times with boiling dichloromethane. There was observed almost no change in the amount of the ionic compound carried, which confirmed that the ionic compound and the carrier bonded to each other with sufficient strength.

Example 5:

25 Preparation of Components for Olefin Polymerization Catalyst from the Compound of Example 2

[0107] An olefin polymerization catalyst component was prepared in the same manner as in Example 4 except that the compound of Example 2 was used as the ionic compound having a carrier bondable functional group. The amount of the ionic compound carried was 0.27 (mmol of ionic compound/g of catalyst component).

30 Example 6:

Preparation of Components for Olefin Polymerization Catalyst from the Compound of Example 3

35 **[0108]** An olefin polymerization catalyst component was prepared in the same manner as in Example 4 except that the compound of Example 3 was used as the ionic compound having a carrier bondable functional group. The amount of the ionic compound carried was 0.23 (mmol of ionic compound/g of catalyst component).

40 Comparative Example 2:

Preparation of Components for Olefin Polymerization Catalyst from the Compound of Comparative Example 1

45 **[0109]** An olefin polymerization catalyst component was prepared in the same manner as in Example 4 except that the compound of Comparative Example 1 was used as the ionic compound having a carrier bondable functional group. The amount of the ionic compound carried was 0.17 (mmol of ionic compound/g of catalyst component).

Comparative Example 3:

Preparation of Comparative Olefin Polymerization Catalyst Component

50 **[0110]** A comparative olefin polymerization catalyst component was prepared in the same manner as in Example 4 except that N,N-dimethylanilinium tetrakis(pentafluorophenyl)borate was used in place of the ionic compound having a carrier bondable functional group. The amount of the ionic compound carried was less than 0.005 (mmol of ionic compound/g of catalyst component).

Example 7:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Example 4 and Olefin Polymerization

[Preparation of Catalyst]

[0111] 4 ml of a solution containing 0.5 mmol of rac-ethylene-bisindenylzirconium dichloride (hereafter, abbreviated as "EBIZ")/liter of toluene was contacted with 0.2 ml of a solution of 0.5 mol of triisobutylaluminum/liter of toluene, to which was added 30 mg of the olefin polymerization catalyst component of Example 4. After stirring the solution for 2 minutes, toluene was removed and 5.0 ml of hexane was added to form a hexane slurry.

[Polymerization of Propylene]

[0112] In a 1.5 liter autoclave were charged 6 ml of a hexane solution of 0.5 mol of triisobutylaluminum/liter /liter of hexane and 8 mol of propylene. After elevating the temperature to 50°C, the above-described slurry type olefin polymerization catalyst was added and polymerization was continued for 60 minutes. The polymerization activity was 85,000 g/g of complex/h, and 2,000 g/g of catalyst/h. Table 1 shows the results obtained. The bulk density of the resulting polymer was as high as 0.40 g/ml, showing no attachment to the wall of the reactor.

Example 8:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Example 4 and Olefin Polymerization

[Preparation of Catalyst]

[0113] 30 mg of the olefin polymerization catalyst component of Example 4 was contacted with 1.0 ml of a solution containing 0.5 mol of triisobutylaluminum/liter of toluene, to which was added 4.0 ml of a solution containing 0.5 mol of EBIZ/liter of toluene. After stirring the solution for 3 minutes, toluene was removed and 5.0 ml of hexane was added to form a hexane slurry.

[Polymerization of Propylene]

[0114] The procedures of Example 7 were repeated except that the above-described olefin polymerization catalyst was used. As a result, granular polymer was obtained. Table 1 shows the results.

[Copolymerization of Propylene with Ethylene]

[0115] In a 5 liter autoclave were charged 6 ml of a hexane solution of 0.5 mol of triisobutylaluminum/liter of hexane and 8 mol of propylene. After elevating the temperature to 50°C, the pressure was raised so that the partial pressure of ethylene reached 9.8×10^4 Pa (1 kg/cm²), and the above-described olefin polymerization catalyst was added and polymerization was continued for 30 minutes. Table 2 shows the results. The polymerization activity was 1.3g/g of complex/h/Pa (130,000 g/g of complex/h/atm), and 10,000 g/g of catalyst/h. The resulting polymer was granular and had a bulk density as high as 0.35 g/ml, showing no attachment to the wall of the reactor.

[Polymerization of Ethylene]

[0116] In a 1.5 liter autoclave were charged 8 ml of a toluene solution of 0.5 mol of triisobutylaluminum/liter of toluene, 100 mg of the olefin polymerization catalyst, and 700 ml of isobutane. After elevating the temperature to 70°C, the pressure was raised so that the partial pressure of ethylene reached 9.8×10^5 Pa (10 kg/cm²), and polymerization was continued for 30 minutes. The polymerization activity was 1.5g/g of complex/h/Pa (150,000 g/g of complex/h/atm), and 1×10^{-2} g/g of catalyst/h/Pa (1,000 g/g of catalyst/h/atm). Table 3 shows the results. The resulting polymer had a bulk density as high as 0.35 g/ml, showing no attachment to the wall of the reactor.

Example 9:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Example 4 and Olefin Polymerization

[Preparation of Catalyst]

[0117] An olefin polymerization catalyst was prepared in the same manner as in Example 8 except that removal of the supernatant, washing with toluene, and slurry formation with hexane were not carried out.

[Polymerization of Propylene]

[0118] In a 1.5 liter autoclave were charged 1.0 ml of a hexane solution of 0.1 mol of n-butyllithium/liter of hexane and 8 mol of propylene. After elevating the temperature to 50°C, the above-described olefin polymerization catalyst was added and polymerization was continued for 60 minutes. Table 1 shows the results. The polymer was granular and showed no attachment to the wall of the reactor.

[Copolymerization of Propylene with Ethylene]

[0119] In a 1.5 liter autoclave were charged 1.0 ml of a hexane solution of 0.1 mol of n-butyllithium/liter of hexane and 8 mol of propylene. After elevating the temperature to 50°C, the pressure was raised so that the partial pressure of ethylene reached 9.8×10^4 Pa (1.0 kg/cm²), and the above-described olefin polymerization catalyst was added and polymerization was continued for 30 minutes. Table 2 shows the results. The resulting polymer was granular and showed no attachment to the wall of the reactor.

[Polymerization of Ethylene]

[0120] In a 1.5 liter autoclave were charged 1.0 ml of a hexane solution of 0.1 mol of n-butyllithium/liter of hexane and 700 ml of isobutane. After elevating the temperature to 70°C, the pressure was raised so that the partial pressure of ethylene reached 9.8×10^5 Pa (10.0 kg/cm²), and polymerization was continued for 60 minutes. Table 3 shows the results. The resulting polymer was granular and showed no attachment to the wall of the reactor.

Example 10:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Example 5 and Olefin Polymerization

[Preparation of Catalyst]

[0121] An olefin polymerization catalyst was prepared in the same manner as in Example 8 except that the olefin polymerization catalyst component of Example 5 was used.

[Polymerization of Propylene]

[0122] The propylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 1 shows the results.

[Copolymerization of Propylene and Ethylene]

[0123] The procedures of copolymerization of propylene and ethylene described in Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 2 shows the results.

[Polymerization of Ethylene]

[0124] The ethylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 3 shows the results.

Example 11:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Example 6 and Olefin Polymerization

5 [Preparation of Catalyst]

[0125] An olefin polymerization catalyst was prepared in the same manner as in Example 8 except that the olefin polymerization catalyst component of Example 6 was used.

10 [Polymerization of Propylene]

[0126] The propylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 1 shows the results.

15 [Copolymerization of Propylene and Ethylene]

[0127] The procedures of copolymerization of propylene and ethylene described in Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 2 shows the results.

20 [Polymerization of Ethylene]

[0128] The ethylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 3 shows the results.

25 Example 12:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Example 4 and Olefin Polymerization

30 [Preparation of Catalyst]

[0129] An olefin polymerization catalyst was prepared in the same manner as in Example 8 except that racisopropylidene(3-t-butylcyclopentadienyl){1-(3-t-butylindenyl)}zirconium dichloride (hereafter, abbreviated as "CTITZ") was used instead of EBIZ.

35 [Polymerization of Propylene]

[0130] The propylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 1 shows the results.

40 [Copolymerization of Propylene and Ethylene]

[0131] The procedures of copolymerization of propylene and ethylene described in Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 2 shows the results.

45 [Polymerization of Ethylene]

[0132] The ethylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 3 shows the results.

50 Example 13:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Example 4 and Olefin Polymerization

55 [Preparation of Catalyst]

[0133] An olefin polymerization catalyst was prepared in the same manner as in Example 8 except that racdimethylsilylenebis{1-(2-methyl-4-benzoindenyl)}zirconium dichloride (hereafter, abbreviated as "2MBIZ") was used instead of EBIZ.

[Polymerization of Propylene]

[0134] The propylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 1 shows the results.

[Copolymerization of Propylene and Ethylene]

[0135] The procedures of copolymerization of propylene and ethylene described in Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 2 shows the results.

[Polymerization of Ethylene]

[0136] The ethylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 3 shows the results.

Example 14:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Example 4 and Olefin Polymerization

[Preparation of Catalyst]

[0137] An olefin polymerization catalyst was prepared in the same manner as in Example 8 except that racdimethylsilylenebis{1-(2-methyl-4-phenylindenyl)}zirconium dichloride (hereafter, abbreviated as "2MPIZ") was used instead of EBIZ.

[Polymerization of Propylene]

[0138] The propylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 1 shows the results.

[Copolymerization of Propylene and Ethylene]

[0139] The procedures of copolymerization of propylene and ethylene described in Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 2 shows the results.

[Polymerization of Ethylene]

[0140] The ethylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 3 shows the results.

Example 15:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Example 4 and Olefin Polymerization

[Preparation of Catalyst]

[0141] An olefin polymerization catalyst was prepared in the same manner as in Example 8 except that racdimethylsilylenebis{1-(2-methyl-4-naphthylindenyl)}zirconium dichloride (hereafter, abbreviated as "2MNIIZ") was used instead of EBIZ.

[Polymerization of Propylene]

[0142] The propylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 1 shows the results.

[Copolymerization of Propylene and Ethylene]

[0143] The procedures of copolymerization of propylene and ethylene described in Example 8 were repeated except

that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 2 shows the results.

[Polymerization of Ethylene]

- 5 [0144] The ethylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used to yield a granular polymer. Table 3 shows the results.

Comparative Example 4:

- 10 Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Comparative Example 3 and Olefin Polymerization

[Preparation of Catalyst]

- 15 [0145] An olefin polymerization catalyst was prepared in the same manner as in Example 8 except that the olefin polymerization catalyst component of Comparative Example 3 was used.

[Polymerization of Propylene]

- 20 [0146] The propylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used. Table 1 shows the results. Almost no polymerization activity was observed.

[Copolymerization of Propylene and Ethylene]

- 25 [0147] The procedures of copolymerization of propylene and ethylene described in Example 8 were repeated except that the above-described olefin polymerization catalyst was used. Table 2 shows the results. Almost no polymerization activity was observed.

[Polymerization of Ethylene]

- 30 [0148] The ethylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used. Table 3 shows the results. Almost no polymerization activity was observed.

Comparative Example 5:

- 35 Preparation of Comparative Olefin Polymerization Catalyst and Olefin Polymerization

[Preparation of Catalyst]

- 40 [0149] 4 ml of a solution of 0.5 mmol of EBIZ/liter of toluene was contacted with 0.2 ml of a solution of 0.5 mol of triisobutylaluminum/liter of toluene, to which was added 2 ml of a solution of 1 mmol of N,N-dimethylanilinium tetrakis (pentafluorophenyl)borate/liter of toluene. After stirring for 2 minutes, 30 ml of toluene and 0.5 g of silica (dried at 450°C for 4 hours) were added. After stirring the solution for 5 minutes, toluene was removed and the residue was washed with hexane and converted into a hexane slurry.

- 45 [Polymerization of Propylene]

- [0150] In a 1.5 liter autoclave were charged 6 ml of a hexane solution of 0.5 mol of triisobutylaluminum/liter of hexane and 8 mol of propylene. After elevating the temperature to 50°C, the above-described olefin polymerization catalyst was added and polymerization was continued for 30 minutes. The polymerization activity was 7,400 g/g of complex/h, and 130 g/g of catalyst/h. Table 1 shows the results obtained. The resulting polymer had a bulk density as high as 0.12 g/ml and was fine powder showing attachment to the wall of the reactor to some extent.

- 55 [Copolymerization of Propylene and Ethylene]

- [0151] The procedures of copolymerization of propylene and ethylene described in Example 8 were repeated except that the above-described olefin polymerization catalyst was used. Table 2 shows the results. The resulting polymer was fine powder showing attachment to the wall of the reactor to some extent.

[Polymerization of Ethylene]

[0152] The ethylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used. Table 3 shows the results.

Comparative Example 6:

Preparation of an Olefin Polymerization Catalyst from the Catalyst Component of Comparative Example 2 and Olefin Polymerization

[Preparation of Catalyst]

[0153] An olefin polymerization catalyst was prepared in the same manner as in Example 8 except that the olefin polymerization catalyst component of Comparative Example 2 was used.

[Polymerization of Propylene]

[0154] The propylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used. A granular polymer was obtained. Table 1 shows the results.

[Copolymerization of Propylene and Ethylene]

[0155] The procedures of copolymerization of propylene and ethylene described in Example 8 were repeated except that the above-described olefin polymerization catalyst was used. A granular polymer was obtained. Table 2 shows the results.

[Polymerization of Ethylene]

[0156] The ethylene polymerization procedures of Example 8 were repeated except that the above-described olefin polymerization catalyst was used. Table 3 shows the results.

Table 1

Polymerization of Propylene				
Example	Activity of Polymerization		Density g/ml	Attachment of Polymer
	gPP/Complex/h	gPP/Catalyst/h		
7	85,000	2,000	0.40	No
8	30,000	600	0.41	No
9	66,000	1,700	0.40	No
10	100,000	7,000	0.43	No
11	62,000	3,300	0.39	No
12	55,000	3,000	0.40	No
13	31,000	1,700	0.40	No
14	20,000	1,500	0.35	No
15	40,000	2,000	0.40	No
C.Ex.4	0	0	-	-
C.Ex.5	7,400	130	0.12	Yes
C.Ex.6	5,000	300	0.37	No

Table 2

Copolymerization of Propylene and Ethylene				
Example	Activity of Polymerization		Density g/ml	Attachment of Polymer
	gPP/mol Complex/h	gPP/g Catalyst/h		
8	130,000	10,000	0.35	No
9	260,000	11,000	0.35	No
10	320,000	29,000	0.35	No
11	160,000	10,000	0.37	No
12	380,000	6,300	0.32	No
13	230,000	13,000	0.33	No
14	310,000	21,000	0.30	No
15	100,000	7,000	0.39	No
C.Ex.4	0	0	-	-
C.Ex.5	30,000	3,000	0.13	Yes
C.Ex.6	20,000	2,000	0.35	No

Table 3: Polymerization of Ethylene

Example	Activity of Polymerization		Density	Attachment
	(gPE/mmol Complex/h/atm) gPE/mmol Complex/h/Pa	(gPE/g Catalyst/h/atm) gPE/g Catalyst/h/Pa	g/ml	of Polymer
8	1.5 (150,000)	1×10^{-2} (1,000)	0.35	No
9	1.0 (100,000)	2×10^{-2} (2,000)	0.32	No
10	0.7 (70,000)	1.2×10^{-2} (1,200)	0.32	No
11	0.75 (75,000)	1.5×10^{-2} (1,500)	0.35	No
12	0.7 (70,000)	0.6×10^{-2} (600)	0.31	No
13	0.7 (70,000)	1×10^{-2} (1,000)	0.31	No
14	0.11 (11,000)	1.3×10^{-2} (1,300)	0.34	No
15	0.6 (60,000)	5.5×10^{-2} (550)	0.35	No
C.Ex.4	0 (0)	0 (0)	-	-
C.Ex.5	0.8 (80,000)	0.2×10^{-2} (200)	0.15	Yes
C.Ex.6	0.08 (8,000)	1.4×10^{-2} (140)	0.33	No

INDUSTRIAL APPLICABILITY

[0157] The ionic compound of this invention, which is used in combination with a carrier, an organometal compound and a Group 4, 5 or 6 transition metal compound, is useful as a catalyst component for olefin polymerization by a slurry process, gas phase process or the like. Using the ionic compound of this invention, catalysts having excellent bonding ability with the carrier can be obtained and it is possible therewith to produce polyolefins having improved powder characteristics and showing no attachment to the inner wall of a reactor.

[0158] The catalyst of this invention is useful for producing a wide variety of polymers from monomers such as ethylene, propylene, 1-butene, 3-methyl-1-butene, 1-hexene, 4-methyl-1-pentene, 1-octene, 1-decene, 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene, 1-eicosene, cyclopentene, cycloheptene, norbornene, 5-methyl-2-norbornene, tetracyclododecene, and further styrene, vinylcyclohexane, vinylcyclohexene, divinylbenzene, dienes, etc.

Claims

1. Ionic compounds represented by general formula (I) below:



(wherein, M is an element belonging to the Group 13 and is preferably Boron;

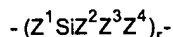
R¹, R² and R³, which may be the same or different, represent each a C₁-C₂₀ hydrocarbonyl group, a substituted hydrocarbonyl group, an alkoxide group, or a halogen atom; Preferably each of R¹, R² and R³ represents a pentafluorophenyl group

R⁴ represents a C₁-C₂₀ alkylene group, a substituted alkylene group, a substituted phenylene group, a silanylene group, a substituted silanylene group, a silalkylene group, a substituted silalkylene group, an oxasilanylene group, a substituted oxasilanylene group, or an oxasilalkylene group, with k, l and m being each 0 or an integer of 1 to 3, and n being an integer of 1 to 4 such that k + l + m + n = 4 and preferably n=1; L is a group represented by general formula (II) or (III) below and is chemically bonded to R⁴:



(wherein R⁵ to R¹², which may be the same or different, represent each a C₁-C₂₀ hydrocarbonyl group, a substituted hydrocarbonyl group, an alkoxide group, or a halogen atom, with at least one of R⁵, R⁶ and R⁷ and at least one of R⁸, R⁹ and R¹⁰ being a halogen atom;

Y is -O-, a C₁-C₂₀ alkylene group, a substituted alkylene group, a phenylene group, a substituted phenylene group, or a group represented by formula:



(wherein Z¹ and Z⁴, which may be the same or different, represent each an alkylene group, a substituted alkylene group, a phenylene group, a substituted phenylene group, -O-, an oxyalkylene group, a substituted oxyalkylene group, an oxyphenylene group, or a substituted oxyphenylene group; Z² and Z³, which may be the same or different, represent each a hydrogen atom, an alkyl group, a substituted alkyl group, a phenyl group, or a substituted phenyl group; r is an integer of at least 1);

when n is 2 or more, each R⁴-L may be the same or different;

D is a monovalent cation selected from the group consisting of carbonium, anilinium, ammonium, ferrocenium, phosphonium, sodium, potassium, or lithium).

2. The ionic compound described in claim 1, wherein L is a halogenated silyl group, a halogenated substituted silyl group, a halogenated silalkyl group, a halogenated substituted silalkyl group, a halogenated oxasilyl group, a

halogenated substituted oxasilyl group, or a halogenated oxasilalkyl group, Preferably a trichlorosilyl group, a methyl dichlorosilyl group, or a dimethyl chlorosilyl group.

3. The ionic compound described in claim 1, wherein R^4 is a substituted phenylene group, and preferably wherein R^4 is a 2,3,5,6-tetrafluorophenylene group.
4. The ionic compound described in claim 1, wherein D is an anilinium ion.
5. A method of producing ionic compounds represented by general formula (I)



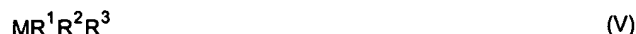
(wherein the symbols in the formula have the same meanings as in claim 1, using compounds defined by (1) to (4) below:

(1) A compound represented by general formula (IV):



(wherein X^1 and X^2 independently represent a hydrogen atom or a bromine atom, and R^4 has the same meaning as in claim 1);

(2) A compound represented by general formula (V):



(wherein M is an element belonging to the Group 13; and

R^1 , R^2 and R^3 , which may be the same or different, represent each a C_1 - C_{20} hydrocarbyl group, substituted hydrocarbyl group, or alkoxide group or a halogen atom);

(3) A compound represented by general formula (VI) or (VII):



(wherein R^5 to R^{12} have the same meanings as in claim 1, R^{13} and R^{14} independently represent a C_1 - C_{20} hydrocarbyl group, substituted hydrocarbyl group, alkoxide group, or a halogen atom); and (4) A halide of a monovalent cation comprising the steps of: (a) reacting a compound defined by (1) with a compound defined by (2); and (b) reacting the product of step (a) with a compound defined by (3); and (c) reacting the product of step (b) with a compound defined by (4).

6. A method of producing ionic compounds represented by general formula (I)



(wherein the symbols in the formula have the same meanings as in claim 1) comprising the steps of:

(a) substituting bromine or hydrogen a compound represented by general formula (IV)



(wherein the symbols in the formula have the same meanings as in claim 5) by lithium with an organic lithium to obtain a lithium substituted compound;

(b) reacting the lithium substituted compound with a Group 13 element containing compound represented by general formula (V)



(wherein the symbols in the formula have the same meanings as in claim 5) to obtain a lithium compound represented by general formula (VIII)



(wherein A is hydrogen or bromine chemically bonded to R^4);

(c) lithionating the compound of general formula (VIII) with an organic lithium and then reacting with a silicon compound represented by general formula (VI) or (VII) below



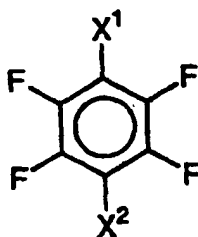
(wherein the symbols in the formulae have the same meanings as in claim 5) to obtain a compound represented by general formula (IX)



and

(d) reacting the compound of general formula (IX) with a halide of a monovalent cation.

7. The method of producing the ionic compounds as described in claim 5 or 6, wherein the compound of general formula (IV) is a compound represented by general formula (IVa)



(IVa)

(wherein the symbols in the formula have the same meanings as in claim 5).

8. A Catalyst component for olefin polymerization comprising the ionic compound described in claim 1 chemically bonded to a carrier.
9. The catalyst component as described in claim 8, wherein the carrier is selected from: a solid having a functional group represented by general formula (X)

-OR

(X)

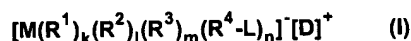
(wherein R is a hydrogen atom, a C₁-C₂₀ alkyl group, alkyl metal or amine, preferably a hydrogen atom), a solid having a hydroxyl group; silica; alumina; or mixtures of silica and alumina.

10. A catalyst for olefin polymerization comprising the following as essential components:

- (a) the catalyst component for olefin polymerization as described in claim 8,
- (b) an organometal compound, and
- (c) a Group 4, 5 or 6 transition metal compound;

Patentansprüche

1. Ionische Verbindungen, wiedergegeben durch die folgende allgemeine Formel (I):



wobei M ein Element gehörend zur Gruppe 13 und vorzugsweise Bor ist,

R¹, R² und R³, die gleich oder verschieden sein können, jeweils eine C₁-C₂₀ Kohlenwasserstoffgruppe, eine substituierte Kohlenwasserstoffgruppe, eine Alkoxidgruppe oder ein Halogenatom darstellen, vorzugsweise jeder der Reste R¹, R² und R³ eine Pentafluorophenylgruppe darstellt,

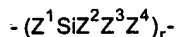
R⁴ eine C₁-C₂₀ Alkylengruppe, eine substituierte Alkylengruppe, eine substituierte Phenylengruppe, eine Silanylengruppe, eine substituierte Silanylengruppe, eine Silalkylengruppe, eine substituierte Silalkylengruppe, eine Oxasilanylengruppe, eine substituierte Oxasilanylengruppe oder eine Oxasilalkylengruppe darstellt, wobei k, l und m jeweils 0 oder eine ganze Zahl von 1 bis 3 sind und n eine ganze Zahl von 1 bis 4 ist, so daß k + l + m + n = 4 und vorzugsweise n = 1 ist,

L eine Gruppe ist, die durch die folgenden allgemeinen Formeln (II) oder (III) wiedergegeben wird, und chemisch an R⁴ gebunden ist:



wobei R⁵ bis R¹², die gleich oder verschieden sein können, jeweils eine C₁-C₂₀ Kohlenwasserstoffgruppe, eine substituierte Kohlenwasserstoffgruppe, eine Alkoxidgruppe oder ein Halogenatom darstellen, wobei mindestens einer der Reste R⁵, R⁶ und R⁷ und mindestens einer der Reste R⁸, R⁹ und R¹⁰ ein Halogenatom ist,

Y -O-, eine C₁-C₂₀ Alkylengruppe, eine substituierte Alkylengruppe, eine Phenylengruppe, eine substituierte Phenylengruppe oder eine Gruppe ist, die durch die Formel wiedergegeben wird:



wobei Z¹ und Z⁴, die gleich oder verschieden sein können, jeweils eine Alkylengruppe, eine substituierte Alkylengruppe, eine Phenylengruppe, eine substituierte Phenylengruppe, -O-, eine Oxyalkylengruppe, eine substituierte Oxyalkylengruppe, eine Oxyphenylengruppe oder eine substituierte Oxyphenylengruppe darstellen, Z² und Z³, welche gleich oder verschieden sein können, jeweils ein Wasserstoffatom, eine Alkylgruppe, eine substituierte Alkylgruppe, eine Phenylgruppe oder eine substituierte Phenylgruppe darstellen,

r eine ganze Zahl von mindestens 1 ist,

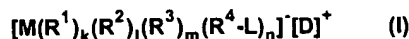
wenn n 2 oder mehr ist, jedes R⁴-L gleich oder verschieden sein kann,

D ein monovalentes Kation ausgewählt aus der Gruppe bestehend aus Carbonium, Anilinium, Ammonium, Ferrocenium, Phosphonium, Natrium, Kalium oder Lithium ist.

2. Ionische Verbindung gemäß Anspruch 1, dadurch gekennzeichnet, daß L eine halogenierte Silylgruppe, eine halogenierte substituierte Silylgruppe, eine halogenierte Silalkylgruppe, eine halogenierte substituierte Silal-

kylgruppe, eine halogenierte Oxasilylgruppe, eine halogenierte substituierte Oxasilylgruppe oder eine halogenierte Oxasilalkylgruppe, vorzugsweise eine Trichlorosilylgruppe, eine Methylchlorosilylgruppe oder eine Dimethylchlorosilylgruppe, ist.

- 5 3. Ionische Verbindung gemäß Anspruch 1, dadurch gekennzeichnet, daß R^4 eine substituierte Phenylengruppe, vorzugsweise eine 2,3,5,6-Tetrafluorphenylengruppe, ist.
4. Ionische Verbindung gemäß Anspruch 1, dadurch gekennzeichnet, daß D ein Aniliniumion ist.
- 10 5. Verfahren zur Herstellung ionischer Verbindungen, die durch die allgemeine Formel (I) wiedergegeben werden,



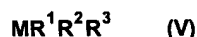
- 15 wobei die Symbole in der Formel die gleichen Bedeutungen wie in Anspruch 1 haben, unter Verwendung der im folgenden durch (1) bis (4) definierten Verbindungen:

(1) Verbindung, wiedergegeben durch die allgemeine Formel (4):



wobei X^1 und X^2 unabhängig ein Wasserstoffatom oder ein Bromatom darstellen und R^4 die gleiche Bedeutung wie in Anspruch 1 hat,

- 25 (2) Verbindung, wiedergegeben durch die allgemeine Formel (5):



- 30 wobei M ein Element der Gruppe 13 ist und R^1 , R^2 , R^3 , die gleich oder verschieden sein können, jeweils eine C_1 - C_{20} Kohlenwasserstoffgruppe, substituierte Kohlenwasserstoffgruppe oder Alkoxidgruppe oder ein Halogenatom darstellen,
- (3) Verbindung, wiedergegeben durch die allgemeinen Formeln (VI) oder (VII):



- 40 wobei R^5 bis R^{12} die gleiche Bedeutung wie in Anspruch 1 haben, R^{13} und R^{14} unabhängig voneinander eine C_1 - C_{20} Kohlenwasserstoffgruppe, substituierte Kohlenwasserstoffgruppe, Alkoxidgruppe oder ein Halogenatom darstellen, und

- (4) ein Halogenid eines monovalenten Kations, aufweisend die Schritte, daß man:
- 45

(a) eine Verbindung definiert durch (1) mit einer Verbindung definiert durch (2) umsetzt,

(b) das Produkt aus Schritt (a) mit einer Verbindung definiert durch (3) umsetzt und

- 50 (c) das Produkt aus Schritt (b) mit einer Verbindung definiert durch (4) umsetzt.

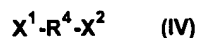
6. Verfahren zur Herstellung ionischer Verbindungen, die durch die allgemeine Formel (I) wiedergegeben werden,



wobei die Symbole in der Formel die gleichen Bedeutungen wie in Anspruch 1 haben,

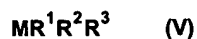
aufweisend die Schritte, daß man:

(a) Brom oder Wasserstoff einer Verbindung, die durch die allgemeine Formel (4) wiedergegeben wird

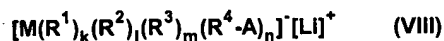


wobei die Symbole in der Formel die gleiche Bedeutung wie in Anspruch 5 haben, mit einem organischen Lithium durch Lithium substituiert, um eine Lithium-substituierte Verbindung zu erhalten,

(b) die Lithium-substituierte Verbindung mit einer Verbindung enthaltend ein Gruppe 13-Element umgesetzt, die durch die allgemeine Formel (V) wiedergegeben wird:

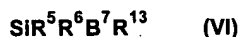


wobei die Symbole in der Formel die gleiche Bedeutung wie in Anspruch 5 haben, um eine Lithiumverbindung zu erhalten, die durch die allgemeine Formel (VIII) wiedergegeben wird

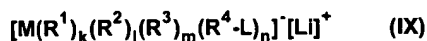


wobei A Wasserstoff oder Brom ist, die chemisch an R^4 gebunden sind,

(c) die Verbindung der allgemeinen Formel (VIII) mit organischem Lithium lithiiert und dann mit einer Silikonverbindung umgesetzt, die durch die allgemeine Formel (VI) oder (VII) wiedergegeben wird,



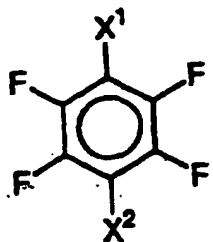
wobei die Symbole in den Formeln die gleiche Bedeutung wie in Anspruch 5 haben, um eine Verbindung zu erhalten, die durch die allgemeine Formel (IX) wiedergegeben wird,



und

(d) die Verbindung der allgemeinen Formel (IX) mit einem Halogenid eines monovalenten Kations umgesetzt.

7. Verfahren zur Herstellung ionischer Verbindungen gemäß Anspruch 5 oder 6, **dadurch gekennzeichnet, daß** die Verbindung der allgemeinen Formel (IV) eine Verbindung ist, die durch die allgemeine Formel (IVa) wiedergegeben wird,



(IVa)

wobei die Symbole in der Formel die gleiche Bedeutung wie in Anspruch 5 haben.

8. Katalysatorkomponente für die Olefin-Polymerisation aufweisend die ionische Verbindung gemäß Anspruch 1 chemisch gebunden an einen Träger.
9. Katalysator Komponente gemäß Anspruch 8, **dadurch gekennzeichnet, daß** der Träger ein Feststoff mit einer funktionellen Gruppe, die durch die allgemeine Formel (X) wiedergegeben wird,



wobei R ein Wasserstoffatom, eine C₁-C₂₀ Alkylgruppe, Metallalkyl oder Amin, vorzugsweise ein Wasserstoffatom, ist, ein Feststoff mit einer Hydroxylgruppe, Siliziumdioxid, Aluminiumoxid oder Mischungen aus Siliziumdioxid und Aluminiumoxid ist.

10. Katalysator für die Olefin-Polymerisation aufweisend die folgenden wesentlichen Komponenten:

- (a) eine Katalysatorkomponente für die Olefin-Polymerisation gemäß Anspruch 8,
- (b) eine Organo-Metallverbindung und
- (c) eine Gruppe 4, 5 oder 6-Übergangsmetallverbindung, vorzugsweise ein Metallocen.

Revendications

1. Composés ioniques représentés par la formule générale (I) ci-dessous :



dans laquelle

- M est un élément appartenant au groupe 13 et est de préférence le bore ;
- R¹, R² et R³, qui peuvent être identiques ou différents, représentent chacun un groupe hydrocarboné en C₁-C₂₀, un groupe hydrocarboné substitué, un groupe alcoxy ou un atome d'halogène, de préférence R¹, R² et R³ représentant chacun le groupe pentafluorophényle ;
- R⁴ représente un groupe alkylène en C₁-C₂₀, un groupe alkylène substitué, un groupe phénylène substitué, un groupe silanylène, un groupe silanylène substitué, un groupe silalkylène, un groupe silalkylène substitué, un groupe oxasilanylène, un groupe oxasilanylène substitué ou un groupe oxasilalkylène,
- k, l et m étant chacun 0 ou un nombre entier allant de 1 à 3, et
- n étant un nombre entier allant de 1 à 4,

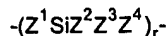
de sorte que k + l + m + n = 4, et de préférence n = 1 ;

L est un groupe représenté par la formule générale (II) ou (III) ci-dessous et est lié chimiquement à R⁴ :



R⁵ à R¹², qui peuvent être identiques ou différents, représentant chacun un groupe hydrocarboné en C₁-C₂₀, un groupe hydrocarboné substitué, un groupe alcoxy ou un atome d'halogène, au moins l'un des radicaux R⁵, R⁶ et R⁷ et au moins l'un des radicaux R⁸, R⁹ et R¹⁰ étant un atome d'halogène ;

Y est -O-, un groupe alkylène en C₁-C₂₀, un groupe alkylène substitué, le groupe phénylène, un groupe phénylène substitué ou un groupe représenté par la formule :



dans laquelle Z¹ et Z⁴, qui peuvent être identiques ou différents, représentent chacun un groupe alkylène, un groupe alkylène substitué, le groupe phénylène, un groupe phénylène substitué, -O-, un groupe oxyalkylène, un groupe oxyalkylène substitué, le groupe oxyphénylène ou un groupe oxyphénylène substitué ;
Z² et Z³, qui peuvent être identiques ou différents, représentent chacun un atome d'hydrogène, un groupe alkyle, un groupe alkyle substitué, le groupe phényle ou un groupe phényle substitué ;
r est un nombre entier égal au moins à 1 ;

lorsque n est égal à 2 ou plus, les groupes R⁴-L peuvent être identiques ou différents ;

D est un cation monovalent choisi dans l'ensemble constitué par les cations carbonium, anilinium, ammonium, ferrocénium, phosphonium, sodium, potassium et lithium.

2. Composé ionique selon la revendication 1, dans lequel L est un groupe silyle halogéné, un groupe silyle halogéné substitué, un groupe silalkyle halogéné, un groupe silalkyle halogéné substitué, un groupe oxasilyle halogéné, un groupe oxasilyle halogéné substitué ou un groupe oxasilalkyle halogéné, de préférence le groupe trichlorosilyle, méthyldichlorosilyle ou diméthylchlorosilyle.

3. Composé ionique selon la revendication 1, dans lequel R⁴ est un groupe phénylène substitué, et de préférence dans lequel R⁴ est le groupe 2,3,5,6-tétrafluorophénylène.

4. Composé ionique selon la revendication 1, dans lequel D est l'ion anilinium.

5. Procédé de préparation de composés ioniques représentés par la formule générale (I)



(les symboles dans la formule ayant les mêmes significations que dans la revendication 1), à l'aide des composés définis en (1) à (4) ci-dessous :

(1) un composé représenté par la formule générale (IV) :



dans laquelle X¹ et X² représentent indépendamment un atome d'hydrogène ou de brome, et R⁴ a la même signification que dans la revendication 1 ;

(2) un composé représenté par la formule générale (V) :



dans laquelle M est un élément appartenant au groupe 13 ; et

R¹, R² et R³, qui peuvent être identiques ou différents, représentent chacun un groupe hydrocarboné en C₁-C₂₀, un groupe hydrocarboné substitué, un groupe alcoxy ou un atome d'halogène ;

(3) un composé représenté par la formule générale (VI) ou (VII) :





formules dans lesquelles R^5 à R^{12} ont les mêmes significations que dans la revendication 1, R^{13} et R^{14} représentent indépendamment un groupe hydrocarboné en C_1 - C_{20} , un groupe hydrocarboné substitué, un groupe alcoxy ou un atome d'halogène ;

(4) un halogénure d'un cation monovalent,

comprenant les étapes suivantes : (a) mise en réaction d'un composé défini en (1) avec un composé défini en (2), et (b) mise en réaction du produit de l'étape (a) avec un composé défini en (3), et (c) mise en réaction du produit de l'étape (b) avec un composé défini en (4).

6. Procédé de préparation de composés ioniques représentés par la formule générale (I)



(les symboles dans la formule ayant les mêmes significations que dans la revendication 1), comprenant les étapes suivantes :

(a) remplacement d'un atome de brome ou d'hydrogène d'un composé représenté par la formule générale (IV)



(les symboles dans la formule ayant les mêmes significations que dans la revendication 5), par du lithium, à l'aide d'un composé organolithié, pour l'obtention d'un composé lithié substitué ;

(b) mise en réaction du composé lithié substitué avec un composé contenant un élément du groupe 13, représenté par la formule générale (V)



(les symboles dans la formule ayant les mêmes significations que dans la revendication 5), pour l'obtention d'un composé lithié représenté par la formule générale (VIII)



(dans laquelle A est un atome d'hydrogène ou de brome lié chimiquement à R^4);

(c) lithiation du composé de formule générale (VIII) avec un composé organolithié et ensuite réaction avec un composé silicié représenté par la formule générale (VI) ou (VII) ci-dessous



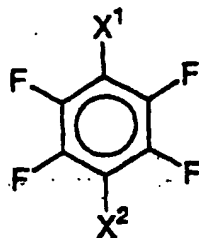
(les symboles dans les formules ayant les mêmes significations que dans la revendication 5), pour l'obtention d'un composé représenté par la formule générale (IX)



et

(d) mise en réaction du composé de formule générale (IX) avec un halogénure d'un cation monovalent.

7. Procédé de préparation de composés ioniques selon la revendication 5 ou 6, dans lequel le composé de formule générale (IV) est un composé de formule générale (IVa)



(IVa)

(les symboles dans la formule ayant les mêmes significations que dans la revendication 5).

8. Composant catalytique pour la polymérisation d'oléfines, comprenant le composé ionique selon la revendication 1, chimiquement lié à un support.

9. Composant catalytique selon la revendication 8, dans lequel le support est choisi parmi :

un solide comportant un groupe fonctionnel représenté par la formule générale (X)

-OR

(X)

dans laquelle R est un atome d'hydrogène, un groupe alkyle en C₁-C₂₀, un composé de type alkylmétal ou une amine, de préférence un atome d'hydrogène ;

un solide comportant un groupe hydroxy ;

la silice ;

l'alumine ;

et des mélanges de silice et d'alumine.

10. Catalyseur pour la polymérisation d'oléfines, comprenant comme composants essentiels les composants suivants :

(a) le composant catalytique pour la polymérisation d'oléfines selon la revendication 8,

(b) un composé organométallique, et

(c) un composé contenant un métal de transition du groupe 4, 5 ou 6, de préférence un métallocène.

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